



PATENT
Docket No. 201835/1020 (BN-3951)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : Thomas M. Clere
Serial No. : RCE of 09/724,158
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For : METHOD FOR MAKING HIGH THERMAL
DIFFUSIVITY BORON NITRIDE POWDERS

Examiner:
Karl Group

Art Unit:
1755

DECLARATION OF THOMAS M. CLERE UNDER 37 C.F.R. § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450
Box:

Dear Sir:

I, THOMAS M. CLERE, declare:

1. I received a Bachelor of Science in Chemical Engineering from Clarkson College of Technology in 1976 and a Masters of Science in Chemical Engineering from Cleveland State University in 1980.
2. I am the Technical and Manufacturing Manager of the Saint-Gobain Advanced Ceramics Structural Ceramics Group, Boron Nitride. I am a member of the American Institute of Chemical Engineers and the Society of Plastic Engineers. I am a former member of the American Ceramic Society and the Electrochemical Society.
3. I am the sole inventor of the above-identified patent application.
4. It is my understanding that the U.S. Patent and Trademark Office has rejected claims 13-16 of my above-identified patent application as being anticipated by or for obviousness over U.S. Patent No. 5,898,009 to Shaffer et al. ("Shaffer I"), U.S. Patent No. 6,048,511 to Shaffer et al. ("Shaffer II"), U.S. Patent No. 5,985,228 to Corrigan et al.

- 2 -

("Corrigan"), and U.S. Patent No. 5,854,155 to Kawasaki et al. ("Kawasaki"). I am submitting this declaration to describe the disclosure of Shaffer I, Shaffer II, Corrigan, and Kawasaki and to show that treatment of boron nitride in accordance with the disclosure of Shaffer I, Shaffer II, Corrigan, or Kawasaki will not achieve a boron nitride powder having a thermal diffusivity of from about $0.15 \text{ cm}^2/\text{s}$ to about $0.20 \text{ cm}^2/\text{s}$, as claimed in my present application.

Shaffer I and II

5. Shaffer I and II, which have the same disclosure, relate to a method of forming pellets or agglomerates of high density boron nitride (BN) by crushing high purity hexagonal boron nitride (hBN) into BN particles extending over a size range of at least $100 \mu\text{m}$ with a majority of the particles having a particle size above $50 \mu\text{m}$ and cold pressing the crushed particles into a compacted form (see Abstract). The compacted form is then granulated and again cold pressed to form pellets or agglomerates of BN particles, which can then be crushed into a powder (see Abstract).

6. In particular, in Example 1, Shaffer I and II teach that high purity BN was crushed to give a crushed powder having an oxygen content of 0.426 %, a surface area of $2.51 \text{ m}^2/\text{g}$, a tap density of 0.92 g/cc , and 0.09 % soluble borates (see Shaffer I at col. 4, lines 45-48 and Table II). The crushed powder had a particle size ranging from about $44 \mu\text{m}$ (-325 mesh) to greater than $420 \mu\text{m}$ (+40 mesh) (Shaffer I, Table II). These references then teach that the crushed powder was compacted using a horizontal press at a pressure of 19,000 psi (Shaffer I, col. 4, lines 48-49). The compacted pieces were then granulated by forcing the material through a screen with openings of approximately $\frac{1}{2}$ inch (Shaffer I, col. 4, lines 49-51). The granulated particles were again compacted at 19,000 psi giving a resultant product with a density of 1.91 g/cc (Shaffer I, col. 4, lines 51-54). Thus, Shaffer I and II teach: (1) crushing high purity BN; (2) compaction of the crushed BN; (3) granulation of the compacted pieces; and (4) compaction of the granulated particles.

7. Example 2 in Shaffer I and II discloses a similar process as in Example 1 for preparing a compacted BN product, however, the crushed starting powder in Example 2 had an oxygen content of 0.60 %, a surface area of $3.02 \text{ m}^2/\text{g}$, a tap density of 0.89 g/cc , 0.14 % soluble borates, and a particle size ranging from $44 \mu\text{m}$ (-325 mesh) to greater than $420 \mu\text{m}$ (+40 mesh) (see Shaffer I at col. 4, line 65 to col. 5, line 4 and Table II). In addition, four additional

- 3 -

granulation/compaction steps as described in Example 1 were performed (Shaffer I, col. 5, lines 4-5). The resultant compact density was 1.91 g/cc (Shaffer I, col. 5, line 6).

8. Example 3 in Shaffer I and II also discloses the same process as in Example 1 for preparing a compacted BN product, however, the crushed starting powder in Example 3 had an oxygen content of 0.275 %, a surface area of 5.26 m²/g, a tap density of 0.85 g/cc, 0.10 % soluble borates, and a particle size ranging from 44 µm (-325 mesh) to greater than 420 µm (+40 mesh) (see Shaffer I at col. 5, lines 21-32 and Table II). The resultant compact density was 1.89 g/cc (Shaffer I, col. 5, lines 31-32).

9. Example 4 in Shaffer I and II discloses a similar process for preparing a compacted BN product, however, the crushed starting powder in Example 4 had an oxygen content of 0.60 %, a surface area of 3.02 m²/g, a tap density of 0.89 g/cc, 0.14 % soluble borates, and a particle size ranging from 44 µm (-325 mesh) to greater than 420 µm (+40 mesh) (see Shaffer I at col. 5, line 37 to col. 6, line 13 and Table II) (this is the same starting powder as in Example 2). The crushed powder was compacted at 19,000 psi using a uniaxial press, and was then formed into granules of 1/16 inch and finer using a granulator (Shaffer I, col. 6, lines 2-5). The granules were once again compacted at 19,000 psi (Shaffer I, col. 6, lines 5-6). The compacts were crushed using a sawtooth and roll crusher and screened through a 120 mesh screen, resulting in a powder having a tap density of 0.68 m²/g (Shaffer I, col. 6, lines 6-8).

10. In order to determine the thermal diffusivity of a product produced as described in Shaffer I and II, an experiment corresponding to Example 1 (i.e., including the four steps of Example 1) was conducted. In particular, BN was: (1) crushed and milled to -325 mesh with a roll mill to produce high purity hBN having an oxygen content of less than 1%, a surface area of 2 to 4 m²/g, a tap density of approximately 0.28 g/cc, a B₂O₃ content (soluble borate) of less than 0.1%, and a particle size ranging from 44 µm (-325 mesh) to greater than 420 µm (+40 mesh). Although the tap density of the crushed BN in this experiment was lower than that described in Example 1 of Shaffer I and II, this modification would not result in a change in thermal diffusivity. In particular, thermal diffusivity is determined by the structure of the BN on the crystallite level. This structure is not affected by the tap density value. (2) The high purity hexagonal boron nitride was then isopressed at 19,000 psi into a compacted form. Although Shaffer I and II describe the use of a horizontal press or uniaxial press to produce the compacted form, compaction via isopressing would not result in a change in thermal diffusivity. The only

- 4 -

difference would be that less alignment of particles would be achieved through the use of isopressing. The thermal diffusivity of the compacted form was measured by laser flash technique (using an Appollo Laser, Model M22) and was found to be $0.09 \text{ cm}^2/\text{sec}$. (3) The compacted form was then crushed in a roll mill (Model 666 "F" Gran-U-Lizer, Modern Process Equipment, Chicago, IL). Although Shaffer I and II describe granulating the compacted form, the substitution of crushing in a roll mill will not impact the resulting thermal diffusivity, as the structure of the BN on the crystallite level will not be changed by either granulating or crushing. (4) Finally, the crushed particles were isopressed again at 19,000 psi into a compacted form. The thermal diffusivity was measured by laser flash technique (using an Appollo Laser, Model M22) and was found to be $0.10 \text{ cm}^2/\text{sec}$.

11. Example 2 of Shaffer I and II relates to the use of additional cycles of granulation and compaction. Such additional cycles of crushing and compaction will not measurably change the thermal diffusivity of the resulting compact, since crushing and compaction will not change the crystallite structure of the BN. Moreover, the modifications in oxygen content, surface area, tap density, percent of soluble borates, and percentages of particle sizes in within the range of $44 \mu\text{m}$ (-325 mesh) to greater than $420 \mu\text{m}$ (+40 mesh) for the starting crushed BN used in Example 2, as compared to Example 1, would not measurably change the resulting thermal diffusivity value. In particular, these properties do not relate to the crystallite structure of the BN and, therefore, would not change the thermal diffusivity value obtained.

12. With regard to Example 3 of Shaffer I and II, an identical process to that described in Example 1 was used with a starting high purity BN having modifications in oxygen content, surface area, tap density, percent of soluble borates, and percentages of particle sizes in within the range of $44 \mu\text{m}$ (-325 mesh) to greater than $420 \mu\text{m}$ (+40 mesh). As described above, such modifications in these properties would not measurably change the resulting thermal diffusivity value.

13. Finally, with regard to Example 4 of Shaffer I and II, a similar process to that described in Example 1 was used, however, a uniaxial press was used (as opposed to a horizontal press), granules of 1/16 inch and finer were produced using a granulator (as opposed to the use of a screen with $\frac{1}{2}$ inch openings), and an additional crushing step was performed after the second compaction. The use of a different type of press, the production of finer granules, and

- 5 -

an additional crushing step would not impact the resulting thermal diffusivity, as these processes would not impact the structure of the boron nitride on the crystallite level. Moreover, the starting high purity BN used in Example 4 is identical to that used in Example 2, and as described in paragraph 11, would not measurably change the resulting thermal diffusivity value.

14. Accordingly, using the methods described in Shaffer I and II, a thermal diffusivity of $0.10 \text{ cm}^2/\text{s}$ was obtained (see paragraph 10). This is below the value claimed in my present application. In contrast to the disclosure of Shaffer I and II, when applying the very dissimilar methodology described in my present application (i.e., sintering after the first isopressing) to the same starting powder described in paragraph 10, a product having a measured thermal diffusivity of $0.18 \text{ cm}^2/\text{s}$ was produced.

Corrigan

15. Corrigan relates to a method for converting hBN particulates into cubic BN (cBN) particles or agglomerates (see Abstract). This reference discloses that the starting hBN powder can be compressed into a billet form and granulated to give particles of a size larger than those commercially available (col. 3, lines 4-6). In addition, Corrigan discloses that the hBN source powder may be densified or pre-pressed into an agglomerated mass or billet at low temperatures, and the mass or billet is then granulated into agglomerated particles of a desired size (col. 2, lines 48-51). Corrigan also discloses that the hBN particles may be vacuum heated and fired, prior to compression and granulation, to remove volatile impurities (col. 3, lines 18-22). In the Examples, the hBN powder is described as "fine-sized HBN powder . . . vacuum fired in the boron nitride thermal decomposition range . . . to obtain a source powder (col. 6, lines 11-14).

16. No further information regarding the starting hBN material in Corrigan is provided. Accordingly, it is not possible to duplicate the disclosure in Corrigan to determine a thermal diffusivity value of the hBN material disclosed. However, as shown above with regard to the disclosure in Shaffer I and II, boron nitride powders will not necessarily have a thermal diffusivity of from about $0.15 \text{ cm}^2/\text{s}$ to about $0.20 \text{ cm}^2/\text{s}$ as claimed in my present application and sintering of a compacted body is required to obtain such a thermal diffusivity value. Sintering of a compacted body of hBN powder is not disclosed in Corrigan.

Kawasaki

17. Kawasaki relates to melamine borate particles, hBN particles and an hBN powder produced using the melamine borate particles, a resin or rubber composite containing the hBN powder, an hBN sintered body, and methods of making the above products (col. 1, lines 4-14). This reference discloses formation of an hBN powder by mixing melamine borate particles, or a mixture containing them, and a crystallization catalyst and firing the mixture in a non-oxidizing gas atmosphere at from 1700 °C to 2200 °C (col. 7, lines 53-56 and col. 8, lines 28-32; Examples 1 and 3). Kawasaki also discloses a method for producing hBN powder by calcining melamine borate particles, or a mixture containing them, to form amorphous BN or hBN, adding a crystallization catalyst, and firing the resulting mixture (col. 8, lines 32-39; Examples 2, 4, and 5). This reference further distinguishes the hBN particles obtained in accordance with the invention of Kawasaki from conventional hBN particles, such as non-aggregated scaly particles, hBN blocks wherein crystals are undeveloped, granulated particles, or particles obtained by pulverizing an hBN sintered body (col. 5, line 27 to col. 6, line 3).

18. For comparison to the Kawasaki patent, the thermal diffusivity of a melamine and boric acid derived BN powder was investigated. Melamine (46 kg) and boric acid (54 kg) were dry mixed and compacted into briquettes. These briquettes were heated in an inconel tube furnace to 950 °C under a nitrogen atmosphere to form amorphous BN (see Examples 2, 4, and 5 of Kawasaki). This amorphous BN was then heated to 2000 °C in an induction furnace under a nitrogen atmosphere to form hBN. The thermal diffusivity was measured by laser flash technique (using an Appollo Laser, Model M22) and was found to be 0.39 cm²/s. This high thermal diffusivity can be attributed to the high degree of agglomeration and crystallinity formed during processing of the powder above 950 °C.

19. A similar thermal diffusivity value (i.e., greater than 0.20 cm²/s) will be obtained using the analogous method described in Kawasaki. In particular, the material produced by the teachings of Kawasaki, in terms of thermal diffusivity, will not differ from the material produced using melamine and boric acid as described in paragraph 18, because the crystallinity of the BN produced in both cases will be similar. More specifically, in Kawasaki melamine diborate is formed and fired at temperatures of 1700-2100 °C (Examples 1-5, col. 11, line 56 to col. 13, line 34 and Tables 2-6). In Examples 2, 4, and 5 of Kawasaki, the melamine diborate mixture is calcined at 800 °C or 1600 °C to form amorphous boron nitride prior to heating to form hBN. Thus, a different feed material is used in Kawasaki than in the experiment

- 7 -

we conducted as described in paragraph 18. However, the crystal structure of hBN formed at high temperature (above 1600 °C) is independent of the feed materials. This is illustrated by a comparison of x-ray diffraction patterns for the material produced as described in paragraph 18 (Exhibits A and B) and the x-ray diffraction patterns for a material produced using melamine diborate as a starting material (Exhibit C). In particular, Exhibit A shows the x-ray diffraction pattern for the starting material used in the experiment described in paragraph 18. This corresponds to Figure 1(a) of Sato et al., "Microstructural Development with Crystallization of Hexagonal Boron Nitride," J. Material Sci. Letters, 16(10):795-798 (1997) ("Sato") attached as Exhibit C, which relates to a starting BN material produced from melamine diborate. The two x-ray diffraction patterns (Exhibit A and Figure 1(a)) show the same two broad peaks. Exhibit B shows the x-ray diffraction pattern for the resulting hBN produced in the experiment described in paragraph 18. This corresponds to Figure 1(e) of Sato attached as Exhibit C, which relates to a resulting hBN product using melamine diborate as a starting material and heating at 2000 °C. The two x-ray diffraction patterns (Exhibit B and Figure 1(e)) again show the presence of the same peaks. Thus, the crystal structure of the two products is the same, although the starting materials and reaction conditions are different. More specifically, the materials are identical, chemically and structurally at the atomic level, due to their treatment at high temperature (above 1600 °C). The same is true of the hBN produced by the method described in Kawasaki as compared to the hBN produced as described in paragraph 18 -- both products are produced by treatment at high temperature (above 1600 °C). Therefore, properties of the hBN produced in Kawasaki and the hBN produced in the experiment described in paragraph 18, such as crystallinity, thermal diffusivity, and electrical conductivity, will be the same. Moreover, the addition of the crystallization catalysts disclosed in Kawasaki merely impacts the rate of reaction and would not alter the crystal structure, and, therefore, the thermal diffusivity, of the resulting product.

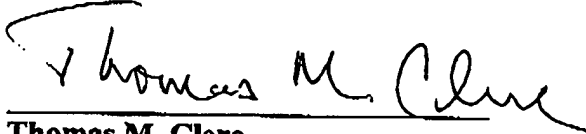
20. Accordingly, hBN with a thermal diffusivity value greater than 0.20 cm²/s will be produced according to the methods of Kawasaki.

21. I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the

- 8 -

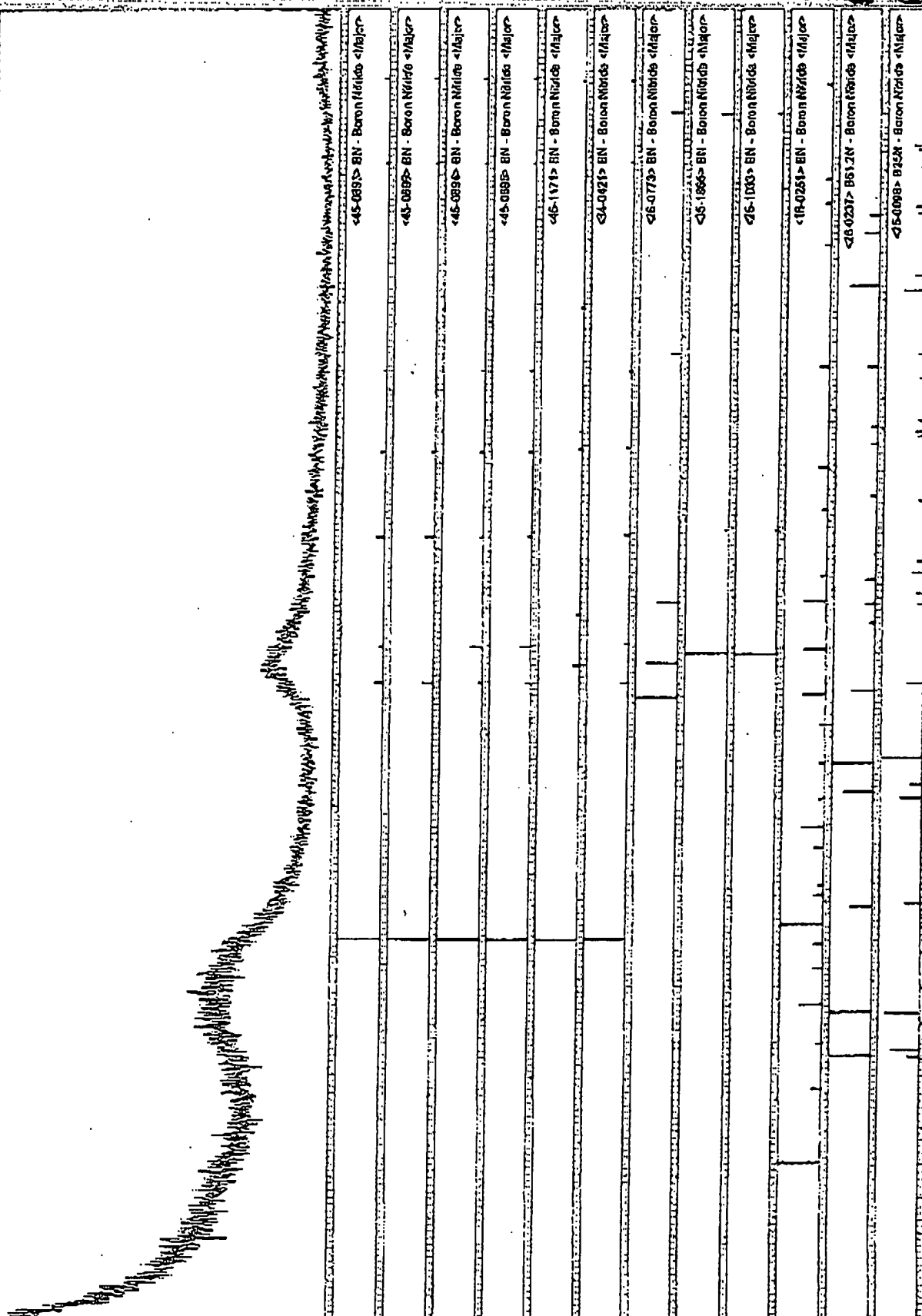
United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: 19-Dec-03


Thomas M. Clere

SCHONCE 14 MDI's 61 AS2866 NITRIDE

Intensity (Counts)



2-theta (deg)

<45-089> BN - Boron Nitride <Major>

<45-089> BN - Boron Nitride <Major>

<45-089> BN - Boron Nitride <Major>

<45-089> BN - Boron Nitride <Major>

<45-171> BN - Boron Nitride <Major>

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<46-186> BN - Boron Nitride <Major>

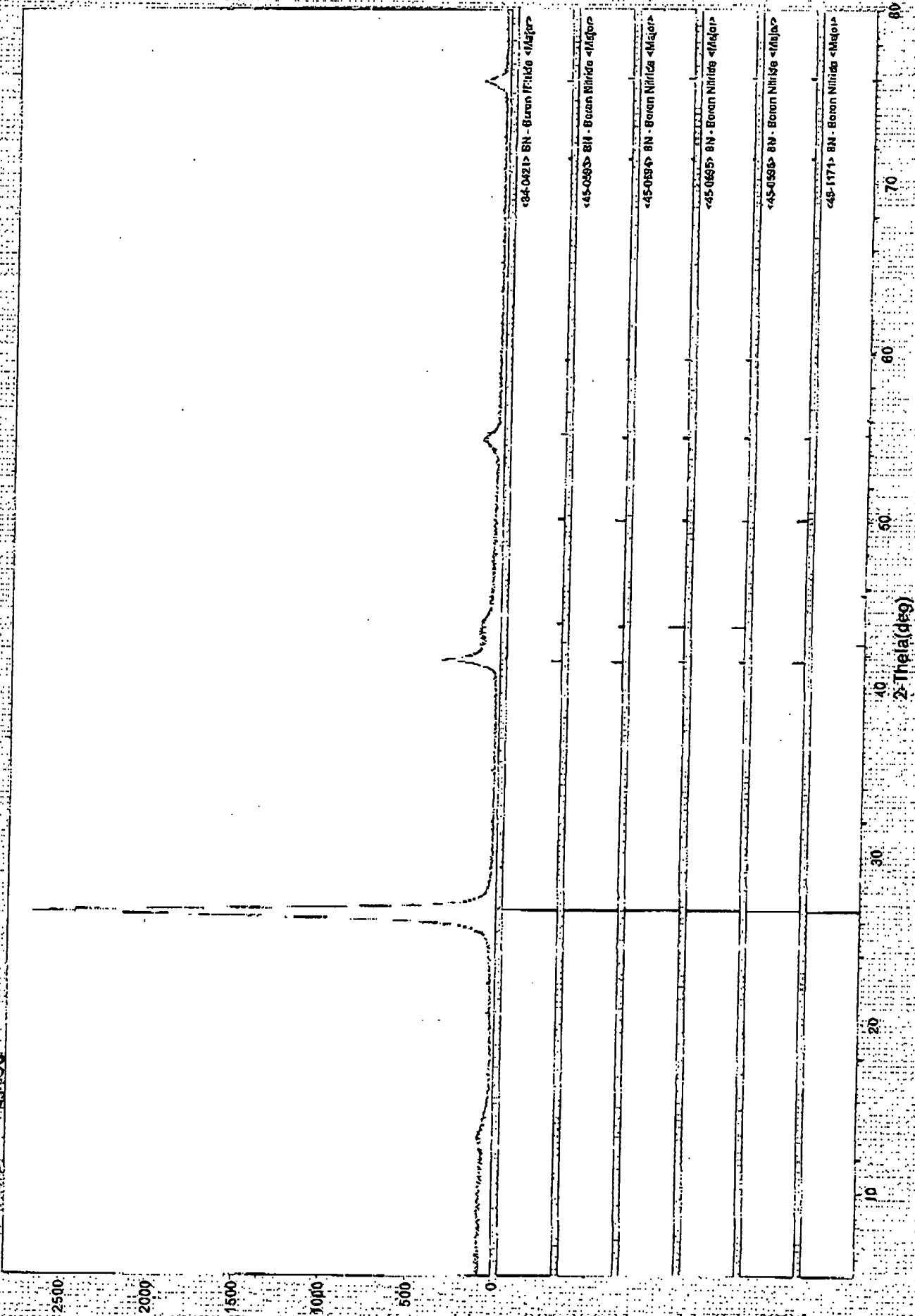
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<46-025> BN - Boron Nitride <Major>

<46-023> BS12N - Boron Nitride <Major>

<45-008> B2N - Boron Nitride <Major>

<CHONG77.MDI> 451350



45-1171 BN - Boron Nitride <Major>

Microstructural development with crystallization of hexagonal boron nitride

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Hexagonal boron nitride can be synthesized by a reaction between a compound containing boron and one containing nitrogen, and numerous production methods combining these have been reported in the literature [1-6]. A turbostratic structure of boron nitride has been produced at a low temperature from a fused urea-boric acid intermediate and ammonia [2]. The term "turbostratic" is used to describe an imperfect structure with no three-dimensional order. It is generally accepted that the turbostratic structure is transformed into an ordered hexagonal form by heat treatment at a temperature above 1500 °C [2, 7, 8], analogous to the carbon graphitization process. The coexistence of boric oxide also influences the rate and extent of the transformation. On the other hand, turbostratic boron nitride can be also produced by thermal decomposition of melamine diborate [9, 10], but its structural transformation at elevated temperatures has not been clarified in detail.

The present letter reports details of the structural transformation with heat treatments of turbostratic boron nitride prepared from melamine diborate, and especially the mode of formation of the grains of boron nitride on the micro-scale.

Turbostratic starting material used in this study was produced by thermal decomposition of melamine diborate in flowing nitrogen at 900 °C, in accordance with the method described in [10]. It was composed of white powders with a columnar shape, in contrast to the cinder-like structure obtained by the urea-boric acid route [2], and it contained about 27 wt% of oxygen as an impurity. The powder specimens were charged into carbon containers lined with BN, and were heated rapidly by induction heating and held at a desired temperature of 1000-2000 °C for 10 min in flowing argon. All of the specimens thus obtained were white powders.

Crystallographic measurements were carried out using an X-ray diffractometer (XRD) RU-2000PL (Rigaku Denki) using a graphite monochromator, and the $d_{(002)}$ spacing and crystallite size $L_{c(002)}$ of BN were calculated from the positions and broadening of the (002) line [2, 6]. Infrared (IR) spectra were measured by the KBr tablet method with an IR-800 spectrometer (Japan spectroscopic). The oxygen impurity content was measured by fusion extraction with a TC-436 (LECO) analyser, and the specific surface area was measured by the Brunauer-Emmett-

Teller (BET) three points method using AUTOSORB-1 (Ionics). Morphological studies were carried out using scanning electron microscopy (SEM) with an ALPHA-50 (Akasi Beam Technology).

Fig. 1 presents typical XRD patterns of the samples obtained at different temperatures. The starting material showed a turbostratic structure, having two broad peaks corresponding to (002) and (10) reflection of BN near 25.6° and near 42.8°, respectively. At 1000 °C the formation of B_2O_3 took place, and this phase remained up to 1900 °C but disappeared completely by vaporization at 2000 °C. Measurements of the galvanic curve indicated that the vaporization of the B_2O_3 started around 1700 °C.

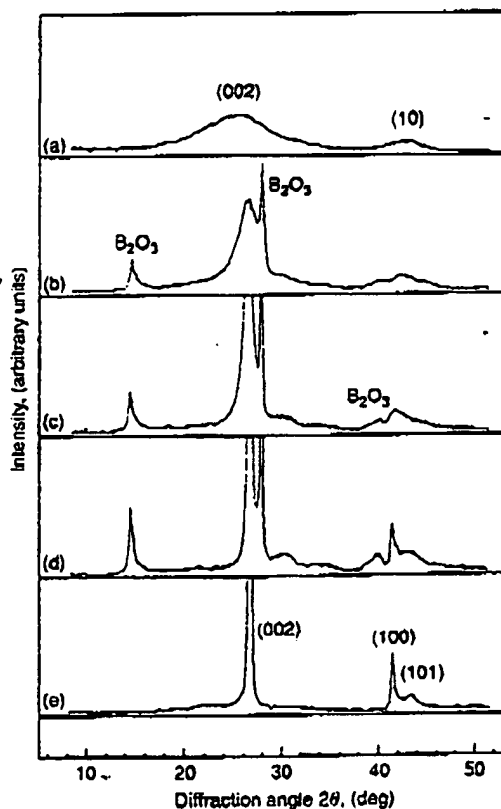


Figure 1 XRD patterns of the samples obtained at different temperatures. (a) starting material, (b) 1000 °C, (c) 1300 °C, (d) 1600 °C, (e) 2000 °C.

and the yield of BN was about 55% at 2000 °C. This suggests that almost all of the oxygen present in the starting materials form B_2O_3 and it vaporizes at elevated temperatures. It was confirmed that the B_2O_3 phase formed was removed by leaching with methanol, consequently, the samples after the treatments were subjected to the following series of experiments.

Fig. 2a presents the oxygen content in the samples obtained at different temperatures. XRD studies indicated that the samples after treatments with methanol exhibited only the BN phase, while they contained an appreciable amount of oxygen (residual oxygen). The raw material contained about 27 wt% of oxygen, and only about half of which was as B_2O_3 at 1000 °C. The residual oxygen level reduced with increasing temperature up to 1600 °C, where part of the decrease may be also in the form of B_2O_3 . When heat-treated above 1700 °C, the residual oxygen content became less than 1 wt%. Fig 2b presents the specific surface area of the samples as a function of heat treatment temperature. The sample obtained at 1000 °C had a high surface area of 250 m²/g, compared to 7 m²/g for the starting one. The magnitude of the surface area reduced with a further increase of temperature, but above 1600 °C its value appeared to saturate at about 10 m²/g. A good linear relationship was also found between the magnitudes of the surface area and the residual oxygen content in the samples, except for the starting one.

Fig. 3 shows the crystallographic changes of BN with increasing treatment temperature. The starting

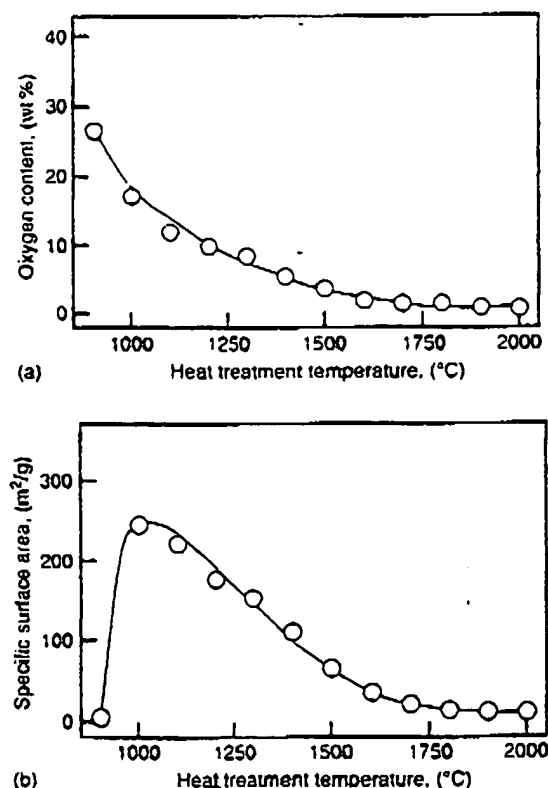


Figure 2 Changes in (a) oxygen contents and (b) specific surface areas of the samples with treatment temperature.

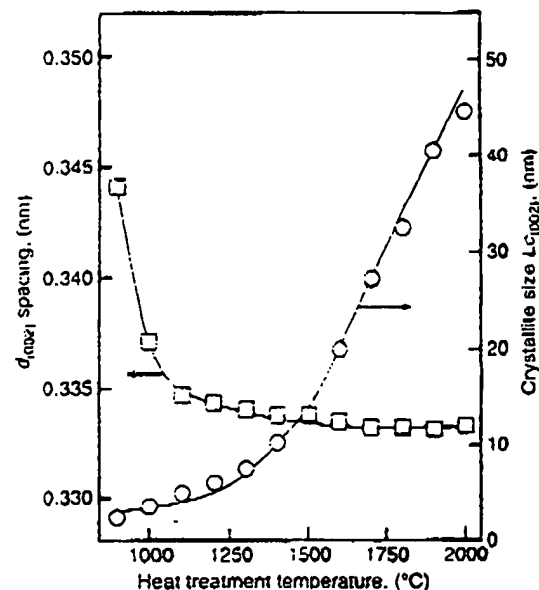


Figure 3 Changes in d_{1002} spacing and crystallite size $L_{c(1002)}$ of boron nitride with treatment temperatures.

materials with turbostratic structures exhibited an elongated $d_{(1002)}$ and a small crystallite size $L_{c(1002)}$, and the heat treatments promoted the ordering of the BN structure. A significant change in the X-ray parameters was observed at which temperature the formation of B_2O_3 took place. With a further increase in temperatures, the $d_{(1002)}$ spacing decreased gradually, but above 1700 °C its value appeared to saturate at the standard 0.333 nm [11]. The heat treatments also caused an increase in crystallite sizes $L_{c(1002)}$, especially at temperatures above 1600 °C. A similar behaviour was also observed for the a -axis parameters, where at 1700 °C the (10) reflection showed a tendency to split into reflections of (100) and (101), indicating an increase in the degree of three-dimensional ordering. On the other hand, the starting materials exhibited mainly two broad IR absorptions near 1400 cm⁻¹ and 800 cm⁻¹ due to BN vibrations [12, 13]. The spectra did not undergo significant changes with increasing temperature, while the absorption peaks appeared to become sharp, and above 1600 °C the maximum position shifted to 1380 cm⁻¹ and that of the latter to 820 cm⁻¹, which may be caused by removal of the strain of the crystal lattice [13]. The crystallization behaviour observed appeared to be similar to that for the turbostratic boron nitride from the urea-boric acid system [2, 7, 8].

Fig. 4 shows the SEM micrographs of the samples obtained at several temperatures. The starting material consisted of columnar particles 30 mm in length and 2 mm in width (Fig. 4a). No significant change in the microstructure was observed up to 1500 °C, while an indistinct island pattern was observed on the surface of the columns. This pattern may be produced by the formation of B_2O_3 and its subsequent removal with methanol. At 1600 °C a drastic change in the microstructure was observed in which large amounts of fine BN grains, ~0.1 mm in

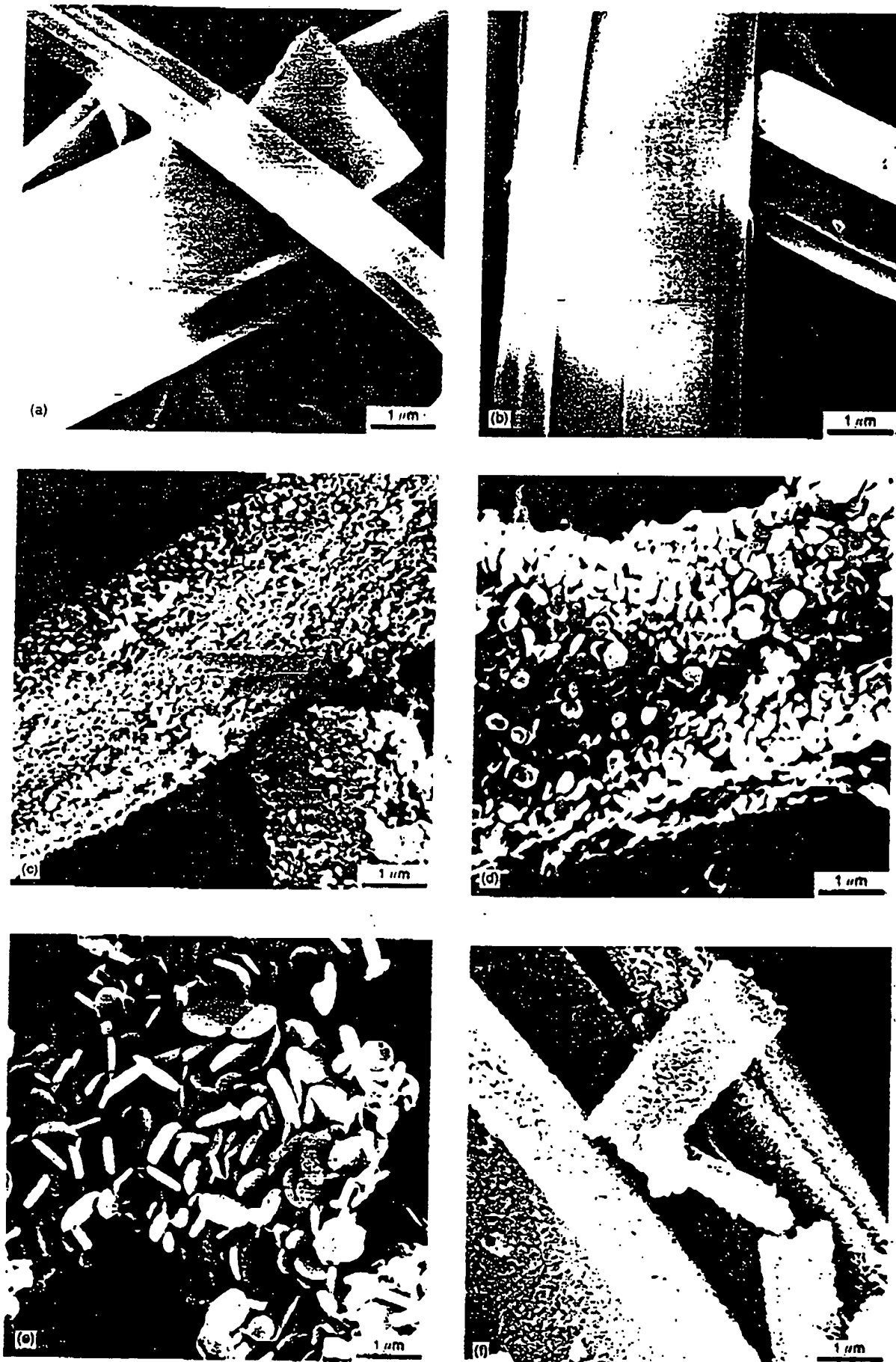


Figure 4 SEM micrographs of the samples obtained at different temperatures. (a) starting material, (b) 1300 °C, (c) 1600 °C, (d) 1800 °C, (e) 2000 °C, (f) 2000 °C reheating of the sample (c) after removing B_2O_3 .

diameter, are generated in the columns (Fig. 4c), and grow with increasing treatment temperature. An exaggerated grain growth took place at temperatures above 1900 °C, in which the spherical form changed into a plate-like one with diameter of the order of one micron (Fig. 4c). This result by SEM observations appears to be consistent with that from XRD and BET measurements. To clarify the effects of B_2O_3 on the growth of the BN grains, the 1600 °C specimen, after treatment with methanol, was reheated at 2000 °C and examined by SEM. The result is shown in Fig. 4f, indicating that the removal of B_2O_3 depressed the growth process on heat treatment. In addition, the crystallite size $L_{(110)2}$ of the resultant samples was 17 nm and the specific surface area was 40 m²/g, which are almost the same as for the sample before reheating. This indicated that the boron oxides formed at low temperatures play an important role in the crystallization or grain growth of BN, as reported by other investigators [5, 14, 15].

The turbostratic boron nitride used in this study is considered to be a B-N-O intermediate product, $n(B_4N_3O_2H)$ as empirical formula, containing a BN skeleton [10]. The turbostratic structure transforms to a partially ordered structure with heat treatment and the value of the $d_{(110)2}$ spacing of BN approaches 0.333 nm at 1600 °C. This suggests that the BN clusters present in the B-N-O intermediate are combined to form a spherical BN particle, as shown in Fig. 4c, and this step may be considered the nucleation and/or nuclear growth of BN. At temperatures above 1700 °C, the partially ordered structure further transforms to a well-crystallized one with three-dimensional ordering, and is accompanied

by grain growth of BN particles, as shown in Fig. 4c. The present work demonstrates the morphological changes involving nuclear and grain growth with the crystallization of turbostratic boron nitride.

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